IMPLEMENTATION CHALLENGES AND PROPOSED SUGGESTIONS FOR NUCLEAR MATERIAL ACCOUNTING MANAGEMENT IN SPENT FUEL REPROCESSING PLANT

HE Li-xia， BAI Lei， MIAO Qiang， YANG Qun

China Institute of Atomic Energy， Beijing, China

ABSTRACT

Nuclear material license holder should develop a nuclear material accounting management system and achieve the closed balance. According to regulations requirements, nuclear material accountancy implementation is based on physical inventory and material measurement. Generally, the spent fuel reprocessing plant operates uninterrupted, annual throughput of nuclear materials is huge, different measurement and analysis techniques are configured combing the reprocessing process. So the closed balance approach of nuclear materials accounting is a great challenge. In order to improve the accuracy and reliability of nuclear material, enhance the timeliness of abnormal detection, near-real time accounting prospect of spent fuel reprocessing plant was researched. In this paper, the crucial aspect affecting the closed balance of nuclear materials was discussed, the factors such as the head-end receiver-shipper difference, the on-line process monitoring accuracy, the applicability of the international target value of nuclear material measurement uncertainty, and also the nuclear material balance model of reprocessing plants. As summarized, proposed suggestions and solutions for nuclear material balance in spent fuel reprocessing plant was put forward on the end of the paper.

Keywords: Nuclear material accounting management; Closed balance approach; Spent fuel reprocessing plant; Near- real time accounting prospect

1. INTRODUCTION

The regulations of China on the nuclear materials control and its implementation guidelines require that the nuclear material license holder shall develop and implement the nuclear material accounting management system. Nuclear material accountancy adopts the closed balance approach. It requires nuclear material in physical inventory must be the measured, and the uncertainty of the measurement system must be well known. Material unaccounted for (MUF) is used for evaluate the facility operation reaches closed balance or not. Suppose σ_{MUF} as MUF value in one accountancy period and σ_{MUF} presents the standard error of material measurement. If $\sigma_{MUF} > 2\sigma_{MUF}$, it can be concluded that the nuclear material didn't reach the closed balance requirement, and there may be nuclear material loss, theft or illegal traffic event has been happed [1] [2] .

Spent fuel is radioactivity, corrosively and chemical toxicity. Spent fuel assembly will be transformed into primary products after chopping, dissolution, co-decontamination, separation, purification, sintered and packaged. The process products changed a lot, nuclear material assemblies transferred from item to bulk, and then to other types of item materials. Uranium and plutonium are also transformed from mixtures to compounds along. Accompany with the process, spent fuel reprocessing plant' s annual throughput is huge, equipment is widely distributed, and process is quite complex. So, implementing nuclear material accountancy in spent fuel reprocessing plant is full of challenges. Measurement accuracy and timeliness are required; integral nuclear material accounting management should be concerned. Indeed, nuclear material accountancy is to be closed.

2. REQUIREMENTS AND IMPLEMENTATION CHALLENGES OF NUCLEAR MATERIAL ACCOUNTING MANAGEMENT IN SPENT FUEL REPROCESSING PLANT

In terms of spent fuel reprocessing capacity,China has formulated a three-step plan,one is to build a 60 ton reprocessing pilot plant every year, the other is to complete the construction of a 200 ton reprocessing plant every year, and the third is to achieve an industrial scale reprocessing capacity of 800 tons every year [3] .The China Atomic Energy Agency (CAEA) issued guidelines for spent fuel of power reactor reprocessing plant nuclear material accountancy HABD-002 / 05 (for trial implementation) in September 2020. The purpose of the guidelines is to recommend compliance with the regulations and implementation rules operation methods and basic procedures related to nuclear material accounting requirements to ensure the rationality and tracking of nuclear material accounting data of spent fuel reprocessing plant and the integrity of technical support documents.

Supposing the operation, it is a big challenge to carry out the nuclear material accounting and management in the spent fuel reprocessing plant. On the one hand, there are many kinds of items involved in the spent fuel reprocessing plant, and it needs to go through a lengthy and complete process from shutdown, emptying and cleaning to restart and operate reasonable. It costs time and budget and generates a lot of waste. The commercial spent fuel reprocessing facilities adopt non-interrupted operation, such as THORP in the UK and UP3 in France [4][5]. On the other hand, the annual throughput of nuclear materials in commercial reprocessing facilities is huge, the MUF value is significant. For example, for the spent fuel reprocessing plant with an annual throughput of 200tHM, the annual throughput of Pu is about 2 tons, if the physical inventory is carried out once a half year. As regulation required , if σ_{MUF} of Pu is less than 20kg, it can be concluded that Pu has reached a closed balance. Obviously MUF of Pu is abundant, and there maybe potential proliferation risk. Therefore, even if the physical inventory of process emptying and equipment cleaning is carried out in the spent fuel reprocessing plant strictly accordance with the provisions of the regulations and implementation rules, the nuclear material has reached a closed balance, the timeliness of nuclear material accounting cannot be realized, and it is not easy to achieve the overall goal of a closed balance accountancy.

3. PROPOSED SOLUTIONS FOR NUCLEAR MATERIALS ACCOUNTING MANAGEMENT IN SPENT FUEL REPROCESSING PLANT

The nuclear material accounting system proposed in HABD-002 / 05 is shown in the figure below, including the trend of nuclear materials MBA and KMP identification. Firstly, the process can be separated into four MBAs according to functional, such as spent fuel assembly receiving, storage and head end-process, chemical process, radioactive waste manage and temporary storage, product storage, etc. Secondly, there are 31 KMPs, including 6 inventories KMPs distributed in spent fuel storage pool, analysis laboratory, radioactive waste management and temporary storage area, uranium plutonium product storage area, etc. Including 14 flow KMPs in the process of spent fuel assembly feeding, spent fuel solution, leached hulls, high radioactive waste, product shipping, etc. And 1 dynamic KMP for plutonium decay loss determination. Thirdly, the corresponding analysis and measurement methods including bulk measurement, chemical analysis (DA) and NDA technology, and it is recommended to quote the international target value of measurement and analysis uncertainty of nuclear safeguards (ITV-2010) as the expected goal of measurement. The guideline mentioned that for the process flow requiring medium-term physical inventory without shut down,t the process monitoring system should be used to measure the important items or batches material, so as to achieve the objective of near- real time data acquisition and processing.

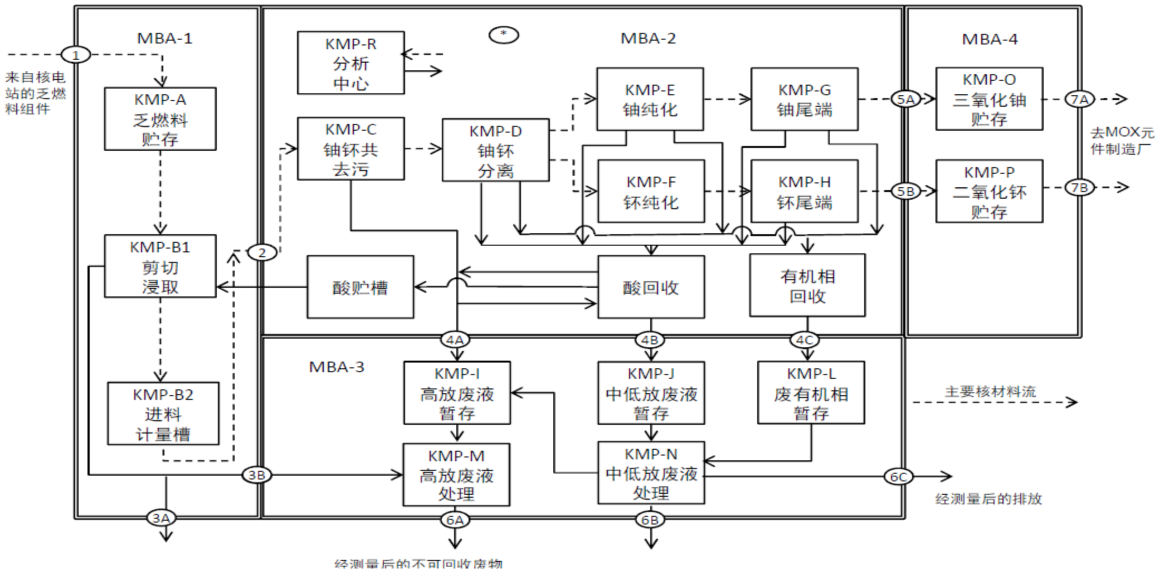

FIGURE 1: NUCLEAR MATERIAL BALANCE AND KEY MEASUREMENT POINT SETTING OF TYPICAL SPENT FUEL REPROCESSING PLANT[1]

The concept of near- real time accounting was originated from 1980s [6] [7] .It is nuclear material accounting management system developed focusing on bulk material processing balance area. It adopts process monitoring technology to achieve the near- real time objective. Based on the nuclear material closed balance approach, MBA is further refined and decomposed according to the characteristics of process and production, process monitoring units and modules are established, appropriate sensors and detectors are used for status monitoring and nuclear material measurement. Then the equipment status and I/O parameters are monitored in real time, setting the near- real time accounting referring to the significant quantity (SQ) of nuclear materials, dynamically analyze the nuclear material inventory, the change of nuclear material stock and the periodic MUF value in the process, and systematically evaluate the monitoring unit, system operation status and nuclear material balance according to the preset index of the process [5][6] [7].Compared with the closed balance accountancy approach, the advantage of near- real time accounting is it can dynamically reflect the operation status of nuclear materials in process components, monitor the quality and changeable of nuclear materials in the process, improve the detection and timeliness of abnormal issues such as leakage, blockage and deviation from expectations, and complement the closed balance accountancy approach. It can well meet the technical requirements for nuclear material accounting in spent fuel reprocessing plant within the regulations and implementation rules.

4. CRUCIAL ASPECTS OF NUCLEA MATERIAL CLOSED ACCOUNTING IN SPENT FUEL REPROCESSING PLANT

Nuclear material accounting in spent fuel reprocessing plant starts from 1AF accountancy tank [8], which is located in MBA1. Its pre-process is spent fuel assembly chopping and dissolution. Leached hulls and high level radioactive waste was produced meanwhile. It is should be emphasized that the nuclear material qualities in these two steps will not matched each other. MBA2 contains complex chemical conversion processes. Uranium and plutonium production will flow to MBA4 for storage after separation, purification and furtherprocess. There are varieties of measurement and analysis methods, and uncertainty is widespread in the data management. In MBA3, it is mainly waste liquid, all of them are flowing KMPs. In MBA4, it is UO_3 and PuO_2 products storage as items.

4.1 Receiver and shipper difference in MBA1

There are three inventories KMPs and four flow KMPs in MBA1. The spent fuel storage is recorded as KMP-A1 and the chopping dissolution cell is recorded as KMP- B1. The spent fuel solution flow to co-decontamination (KMP-C) step in MBA2 for further process; the main components of solid waste are leached hulls and assembly ends, which are recorded as KMP-3A.

Spent fuel assemblies, leached hulls and assembly ends are the most concerned objects of nuclear material accounting and control, which provide basic parameters for processing and accountancy respectively. Nuclear material content in spent fuel assemblies was carried from nuclear power plant. The nuclear material accounting data at KMP-B1 is derived from volume determination and nuclear material concentration measuring. That produces accountancy differences among KMP-A, KMP- B1 and KMP-B2. It definitely contains the receiver and shipper difference, which will not be further discussed in this paper, but it should be concerned that the accuracy of measurement data needs to be reasonably controlled.

In fact, nuclear materials in spent fuel assembly was calculated by computer code, and its accuracy and uncertainty are further corrected and evaluated by using the analysis data of solution. At KMP-B1, specialized NDA equipment and instruments for burnup determine are configured. The data of nuclear and non-nuclear materials in spent fuel solution is collected during the process, the computer model is further corrected and the NDA measurement equipment is consolidated calibrated. The reliability of computer code, the accuracy of NDA method is gradually improved through continuous iterative optimization, so the difference between computer code calculation results and NDA measurement gradually reduced.

4.2 Process monitoring and analysis of nuclear materials in MBA2

In MBA2, to quantify the nuclear material in intermediate accountancy tank is also a challenge. The process is equipped with material liquid buffer and transfer tank to ensure the material transportation continuity. For example, pulse extraction column is usually used in U and Pu co- decontamination and separation process. It works in dynamic operation status. So it is not easy to get the exactly nuclear material by volume method or sampling analysis method. In the purification process, the evaporator also works in continuously, so the nuclear material can't get by DA analysis technical. Generally, it is difficult to sample and analyze U and Pu content in these operating units, they are indirectly estimated by process parameters such as liquid flow ratio, acidity and pulse conditions, and also can be estimated by using mathematical model instead of working parameters [9][10], which were depended to on- line monitoring.

Near-real time accounting needs to optimize the configuration according to the status of the facilities. For the running reprocessing plants, the nuclear materials in process can be monitored by increasing NDA measurement points. It is helpful for improving the timeliness of abnormal event detection, and its disadvantage is measurement uncertainty and operation budget are increased simultaneously. For the newly-built reprocessing plants, it is recommended to prompted nuclear material accounting system in the project's preparation and design stage. The logic optimization is carried out in the commissioning process, and the corresponding parameters can be obtained during its operation. The near-real time accounting and closed balance accountancy evaluation of nuclear materials can be compiled during the facility design, so as to improve the detection ability of abnormal events and feedback the process operation status.

For uranium production (KMP-g) and plutonium production (KMP-h) in MBA2, near-real time balance control units can be configured in parallel. Volume and concentration measurement equipment can be installed in the process containers, supplementary measurement instruments can be installed in the process storage tank as far as possible, flow and concentration measurement instruments can be installed for some liquid flows [11]. For the process areas that cannot be measured directly, the estimation method shall be studied.

4.3 Applicability of international target values 2010 for measurement uncertainties in safeguarding nuclear material (ITV-2010)

HABD-002/05 refers to the recommended ITV-2010, including various random and system uncertainty target values of DA and NDA measurement. These data are derived from the application experience of various facilities, determined and released after evaluation by IAEA and international experts, and recommended to all operating facilities and national nuclear material accounting and control system. Table 1 and table 2 list the nuclear material accounting and analysis method ITV-2010 [12] for spent fuel reprocessing facilities, where u (R) represents random uncertainty and U(s) represents system uncertainty.

TABLE 1: BULK MEASUREMENT UNCERTAINTY AND ITV

Mesurement	Instrument	Uncertainty component (/% rel.) U(r)	U(s)	ITV (/% rel.)
Volume	Electromanometer (Accountability tanks)	0.05	0.1	0.12

	Electromanometer (Process tanks; high concentration)	0.2	0.23	0.28
	Electromanometer (Process tanks; low concentrationn)	1	1	1.4
	Electromanometer (Accountability tanks)	0.3	0.2	0.36

TABLE 2: NUCLEAR MATERIAL ACCOUNTING MEASUREMENT ITV

Material	Item	Instrument	ITV (/% rel)
Dissolver solution	Uranium and plutonium Concentration and isotopes abundance	Isotope dilution mass spectrometry HKED	0.28 0.28
Uranium and plutonium production	Uranium and plutonium concentration	Coulometer Potentiometric titration	0.14 0.21
	Isotopic abundance	Mass spectrometry	0.28
Process Production (Uranium and Plutonium concentration)	High concentration	Potentiometric titration Coulometer K-edge absorption	0.21 0.14 0.42
	Low concentration	Isotope dilution mass spectrometry	0.28
	Liquid waste	X-ray fluorescence	2.8

Liquid volume measurement is crucial factor effecting nuclear material closed balance in spent fuel reprocessing plant. Nowadays, high precision electromanometer is commonly used for the spent fuel assembly dissolution volume measurement. In principle, there is functional relationship between solution density and the liquid height level. By measuring the differential pressure of the liquid height level, the height and the density of the liquid can be determined. The relationship can be calibrated before the tank and equipment run in the process. Measured accuracy depends on the differential pressure. Additionally, at the bottom of tank, the uncertainty will increase while there is solid deposition or air flow rate through the pressure pipe is uneven.

4.4 Closed balance accountancy of nuclear materials

For the nuclear material closed balance accountancy, MUF evaluation model should be researched according to its σ_{MUF}, measuring and analysis technical should also be configured simultaneously. While the measuring and analysis data is feedback and calculate according to the ITV parameters as listed in table 2, then the deviation can be found and targeted MUF value can be estimated and evaluated. If the random and system uncertainty can't be improved and the synthetic uncertainty can't achieve the ITV recommended parameter, the σ_{MUF} won't be optimized. On this way,the technical configuration should be adjusted according to the actual situation and operation appropriately.

For example, in spent fuel reprocessing plant, the near-real time accounting system based on the monitoring unit can be separated into three subsystems. First, the working model is biased towards process and calculation.The process model needs to be developed and optimized according to the monitoring, measuring and corresponding algorithms. The models involving hardware need to take into account the operation, maintenance and equipment layout; the calculation model needs to timely calculate and convert a large number of basic data obtained by monitoring and measuring into data that can reflect process operation status, potential loss of nuclear materials and other abnormal conditions. Second is the key monitoring equipment, analyzing instruments and measuring technical. Based on the reprocessing process model, glove box layout, physical parameters such as instruments and equipment, the number and characteristics of measuring points and other parameters, it is necessary to focus on the applicability of equipment and analysis errors. The third is data management system. A large number of original data are transmitted and reported to the central platform after being processed and analyzed by the calculation model, and integrated to generate operation charts, state curves, near-real time accounting reports and others. It is necessary to focus on the authenticity of the original data, the continuity of data transmission and the adaptability of the integration algorithm.

5. CONCLUSION

Accelerate the development of spent fuel reprocessing, nuclear material accounting management scheme of spent fuel reprocessing plant is the most crucial element of compliance operation. The practical significance lies in the solution of nuclear material accounting and the innovative development of restrictive technical schemes.

Combined with the actual operation and management of spent fuel reprocessing plant, the most practical suggestion for nuclear material accountancy is to adopt the system strategy of combining material closed balance accountancy with near-real time accounting. MUF evaluation executed through MBA, KMP and monitoring unit setting, on-line monitoring and other specific measuring method to meet the requirements of the regulations and implementation rules. Make full use of online monitoring data and operation status to feed back the nuclear material inventory and its change trend in near-real time. Based on that, timely identify abnormal events, interactively update process status and early warning parameters, reduce the system false alarm rate as much as possible, and improve the timeliness of nuclear material accounting in the process of nuclear material production. Statistical methods and probability models are used to make reasonable prediction, and identify the material loss or deposition areas along with the process. When the process is shut down or the facility is decommissioned, it can provide very convenient information for the implementation of detention measurement, decommissioning source item investigation and other needs.

During the planning, design, construction and commissioning of the new spent fuel reprocessing plant, the nuclear material accounting program and measuring technical configuration shall be considered in advanced.That can improve the accuracy, timeliness and efficiency of nuclear material closed accountancy during facility operation. The on- line monitoring and analyzing combing with the critical safety, radiation protection and chemical monitoring programs can provide input information for facility design, construction, commissioning and operation, Promote the design and construction of the nuclear material accounting can help license holder to achieve the highest efficiency cost ratio while realizing the function.

REFERENCES

[1] Regulations of the State Council and the people's Republic of China on the control of nuclear materials, 1987.

[2] National Nuclear Safety Administration, energy administration, commission of science, technology and industry for national defense, detailed rules for the implementation of the regulations of the people's Republic of China on the control of nuclear materials[S], Beijing National Nuclear Safety Administration, 1990.

[3] The development safety of nuclear industry is guaranteed [R], people's daily, 2017-02- 16.

[4] ZHANG Qi, suggestions on accelerating the development of spent fuel reprocessing in nuclear power plants [J], China energy, 2019,1,44-47.

[5] SHI Lei, LI Jinying, HU Yantao. The construction of spent fuel reprocessing plant should be put on the agenda [J], energy, 2018,6,93-96.

[6] T.K.Li, E.A.Hakkila, S.F.Klosterbuer,etal. Evalution and Development Plan of NRTA Measurement Methods for the Rokkasho Reprocessing Plant[C]//36th Annual Meeting. USA:INMM,1995.

[7] HE Lixia，CHENG Yimei, YANG Qun. Concept and Design of Nuclear Material Accounting System in Spent Fuel Reprocessing. Atomic Energy Science and Technology,2020.

[8] IAEA, Detailed Description of an SSAC at the Facility Level for Irradiated Fuel Reprocessing Facilities (STR-193) [M].Vienna, IAEA, 1986.4

[9] BARRY J, BRIAN B, STEVE B, et al. NRTMA: Common purpose: Complementary approaches, IAEA-SM-367/8/03[R]. Vienna: IAEA, 2001.

[10] INDUSI J P, MARCUSE W. Method to determine the minimum cost measurement plan consistent with any feasible limit of error on MUF[C]//Meeting of Institute of Nuclear Materials Management. USA: [s. n.], 1974.

[11] B.B.Cipiti, Optimizing Near Real Time Accountability for Reprocessing[C] //53rd Annual Meeting. USA:INMM, 2012. [12] IAEA. International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials (STR-368) [M]. Vienna , IAEA , 2010.